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Combustion Study of Different Transitional Metal Oxide based on AN/MgAl Composites Gas Generators

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Abstract

Ammonium nitrate (AN)-based composite gas generator have attracted a considerable amount of attention because of the clean burning nature of AN as an oxidizer. However, ammonium nitrate-based gas generator has several major problems, namely, poor ignitability, a low burning rate, low energy, and high hygroscopicity. The addition of different transitional metal oxides and MgAl mechanical alloyed proved to be effective in improving the burning characteristics of AN-based gas generator. In this research work, combustion study of different transition metal oxide based on AN/MgAl composites gas generators was studied. Gas generators were combusted at the pressure of 1 MPa, 3 MPa and 5 MPa in the combustion chamber and the burning rates were determined. It was stated that the addition of metal oxides into the composition of the gas generators improves ignition at low pressure and increases the burning rate. The use of the mechanical MgAl alloys as a fuel allowed the ignition of the gas generator at a lower temperature. The method of thermogravimetric/differential thermal analyzer (TG/DTA) was used to investigate the effect of metal oxides addition on the AN/MgAl-based gas generators thermal decomposition characteristics.

1. Introduction

A composite gas generator is a heterogeneous phase and consists of a synthetic plastic binder matrices made of metals and metal alloys and components crystalline of oxidizers. They are used as fuel in vehicles, strategic missiles, and engines in technical fields.

Composite ammonium perchlorate (AP) is widely used in missile production as an energy fuel as it is quite simple to manufacture, with a particular impulse and characteristics of combustion. However, hydrogen chloride, ions of chlorine and residual chlorinated acids in the composition of the combustion products of fuel based on AP pollute the environment [1]. In the coming years to reduce environmental problems, various research studies will be carried out to produce smokeless combustible energy fuels without chlorine. Many studies support the use of HMX and RDX [2] as an oxidant instead of AP, addition of magnesium [3] into the rocket fuel of AP, the use of ADN [4] and energy fuels based on AN [5–7].

Ammonium nitrate (AN), being cheaper and easily available, is widely used as an oxidizing agent in energy fuels. However, ammonium nitrate has several significant shortcomings such as low burning rate, slow flammability, high hygroscopicity and low energy [8]. Despite these disadvantages, AN is widely used as an oxidizing agent in the energy fuels. Because AN is a natural, pure and chlorine-free combustible. The use of MgAl alloy in AN based gas generator improves its thermodynamic characteristics. Because it is known that the melting point and ignition of MgAl alloy is lower than that of pure metals [9]. This property of MgAl alloys improves the ability of ignition of gas generators on the basis of AN MgAl [10–13]. Many studies have declared that the accession of variables of metal oxides in the composition of the energetic materials on the basis of AN improved combustion characteristics[14–19]. Although some metal

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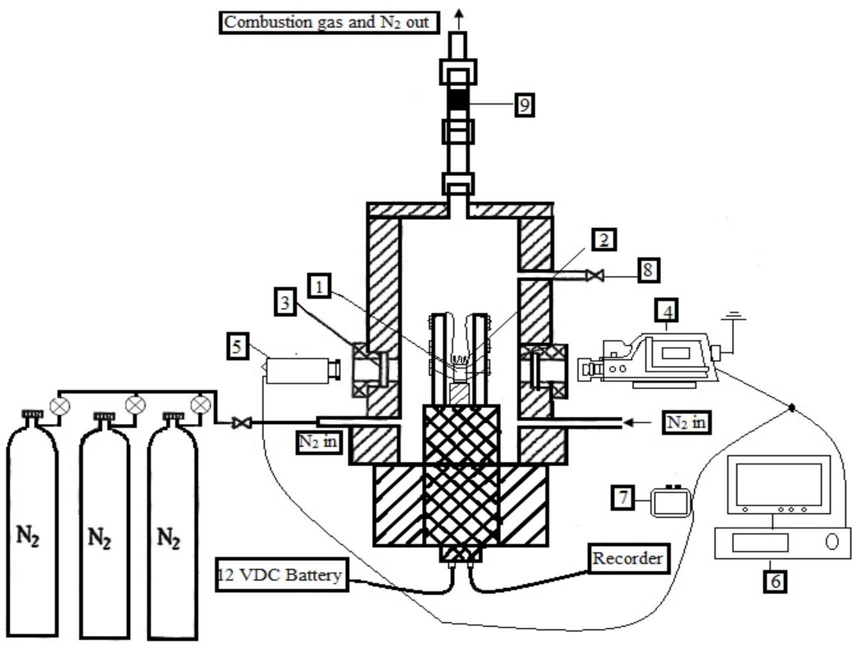


Fig. 1. Scheme of the combustion chamber under pressure: 1 – sample; 2 – nichrome wire; 3 – window; 4 – high-speed video camera; 5 – camera; 6 – transformer; 7 – PC; 8 – system for pressure control; 9 – filter.

oxides exert a negative effect on combustion of mechanical MgAl alloy in the composition of the composite [20]. Energetic gas generators are widely used in space and other technical areas. Therefore, structuring and preparing gas generators is very important. Many studies have been conducted in these areas [21–23].

In this study, the burning characteristics of with various metal oxides (MeOx – TiO_2, Cr_2O_3, ZrO_2) on the AN/MgAl based gas generators were studied.

2. Experimental part

2.1 Materials and propellant samples

Mechanical treatment (5 min) of ammonium nitrate (purity 99%) powder was carried out in a planetary mill. Ammonium nitrate is used as an oxidizer in the condensed mixture with a diameter of 212–250 μm. A mechanical MgAl alloy (50/50) is used as a gas generator its diameter is 50–70 μm. The diameter of the metal oxide particles is 5–6 μm, it acts as a catalyst. Paraffin is used as a binder.

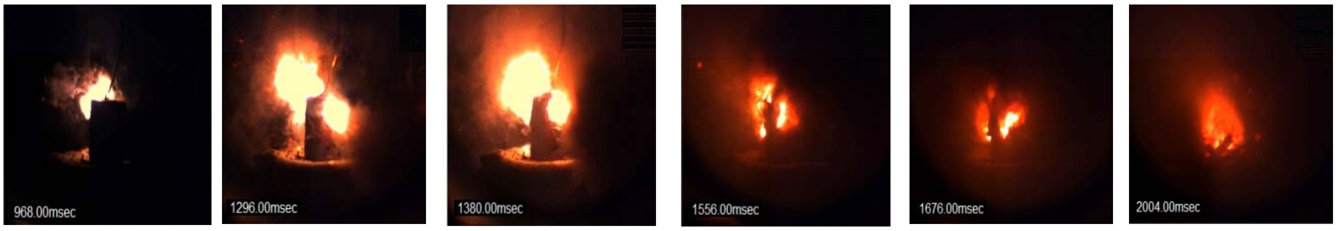
2.2 Measurement of burning characteristics

The diameter of gas generators is 6 mm and the length is 10 mm. Combustion was investigated under gas pressure in the combustion chamber. Each sample was heated up and ignited with nichrome wire. Each of the gas generators is ignited under a pressure of 1–5 MPa. The combustion of gas generators was registered using high-speed cameras. These recorded videos were used to determine the burning rate. The degree of measurement error of 0.01 mm is measured from the dependence of the height of the surface combustion.

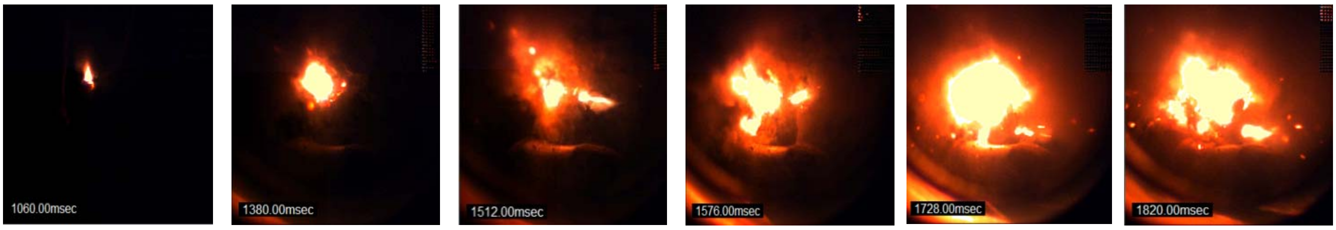
All measurements were made 3 times under pressure and were calculated using the average burning rate. If 1/3 of the gas generator is not ignited or does not burn the burning rate is not determined. Figure 1 shows the equipment for practical work.

2.3 Measurement of thermal decomposition behavior

Thermal analysis is a quick and effective way to study thermal ignition of energetic materials. Characteristics of thermal decomposition are determined and studied by using thermogravimetric/differential thermal analyzer (TG-DTA) in the temperature range of 30–600 °C. The equipment operates in atmospheric pressure in a stream of nitrogen (300 cm^3/min). TG-DTA is working with the heating rate of 20 K/min. Aluminum pans (height 2.5 mm and diameter 5 mm) were used as forms for samples. The sample with the mass 1 mg is placed in thermal connection with aluminum pan. In TG-DTA equipment, for each obtained sample, the standard line of TG-DTA was measured three times.



(AN/MgAl, 3 MPa, time of burning – 824 msec, h = 8.58 mm)



(AN/MgAl, 5 MPa, time of burning – 664 msec, h = 9.86 mm)

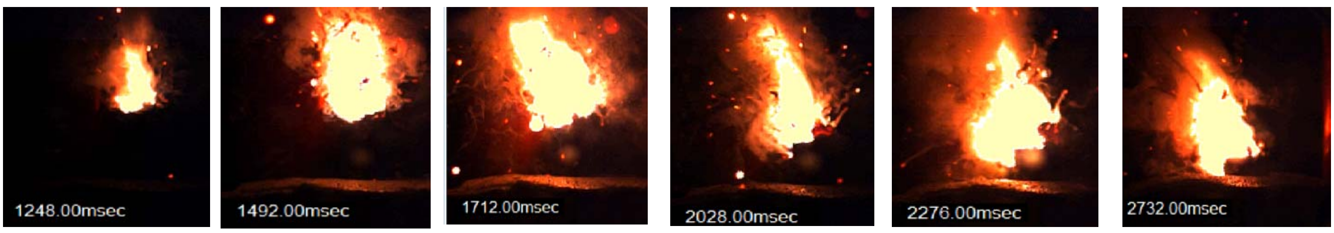
Fig. 2. Burning cinegram of the AN/MgAl – based gas generators at a pressure of 3 MPa and 5 MPa.

3. Results and discussion

Figure 2 shows the mechanism of combustion of AN/MgAl-based gas generators in the combustion chamber at a pressure of 3 MPa and 5 MPa. In Figure 2 shows of the AN/MgAl-based gas generators depending on the increase in pressure the increases in burning rate in the form of a line. Also AN/MgAl-based gas generators do not burn under the pressure of 2 MPa, so the pressure lower than 2 MPa is considered to be the lower ignition limit. The ignition limit is used to improve ignition under low pressure of gas generators. Addition of metal oxides to the system has reduced the flammability limit and provided high speed. The increasing of the flammability limit is due to the disadvantages of ammonium nitrate. In this research work, with a view to reduce these disadvantages, we investigated the mechanisms of combustion by adding the composite transition metal oxide to gas generators on the basis of ammonium nitrate.

Figure 3 shows combustion cinegrams of AN/MgAl/Cr_2O_3 – based gas generator at a pressure of 1 MPa, 3 MPa and 5 MPa.

(AN + MgAl + Cr_2O_3, 1 MPa, time of burning – 788 msec, h = 7.29 mm)



(AN + MgAl + Cr_2O_3, 3 MPa, time of burning – 1028 msec, h = 11.38 mm)

(AN + MgAl + Cr_2O_3, 5 MPa, time of burning – 480 msec, h = 11.31 mm)

Fig. 3. Cinegrams of combustion of gas generator based on AN/MgAl/Cr_2O_3 at 1 MPa, 3 MPa and 5 MPa.

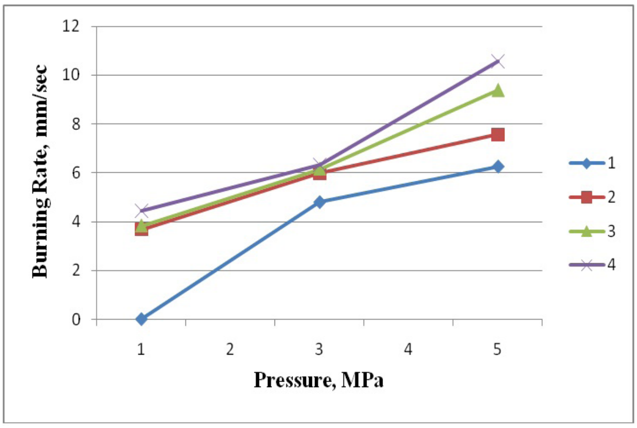


Fig. 4. Graph of the dependence of the burning rate on the pressure of the nitrogen gas: 1 – AN/MgAl; 2 – AN/MgAl/ TiO_2; 3 – AN/MgAl/ZrO_2; 4 – AN/MgAl/Cr_2O_3.

In Fig. 4 shows the characteristics of the burning rate of AN/MgAl/MeOx – based composite gas generators (MeOx – TiO_2, Cr_2O_3, ZrO_2) under different pressures.

In Figs. 3 and 4 shows the increase in the linear burning rate of AN/MgAl – based the gas generators supplemented with metal oxides in comparison with a system AN/MgAl due to the increase of linear pressure and reduction of the lower limit ignition of up to 1 MPa. Also, the increased burning rate of metal oxides showed improve burning characteristics and corrected some disadvantages of ammonium nitrate. According to the obtained experimental results, it can be seen that the gas generators are completely burned with a high combustion rate.

Figure 5 shows the results of thermogravimetric and differential thermal analysis of the AN/MgAl and AN/MgAl/MeOx – based composite gas generators.

In Fig. 5a shows a line of TG-DTA for AN/MgAl – based composite gas generators. The endothermic peaks at the temperature of 58.6 °C and 129.9 °C were identified according to the phase changes of ammonium nitrate and melting point of mechanical MgAl alloy. After that there appeared the exothermic peaks in the temperature range of 136.4–202.5 °C and 245.3–317.0 °C. In addition, there were observed two reduced processes in TG line. The first one was the mass loss in the temperature range from 136.4 °C to 245.3 °C. The second

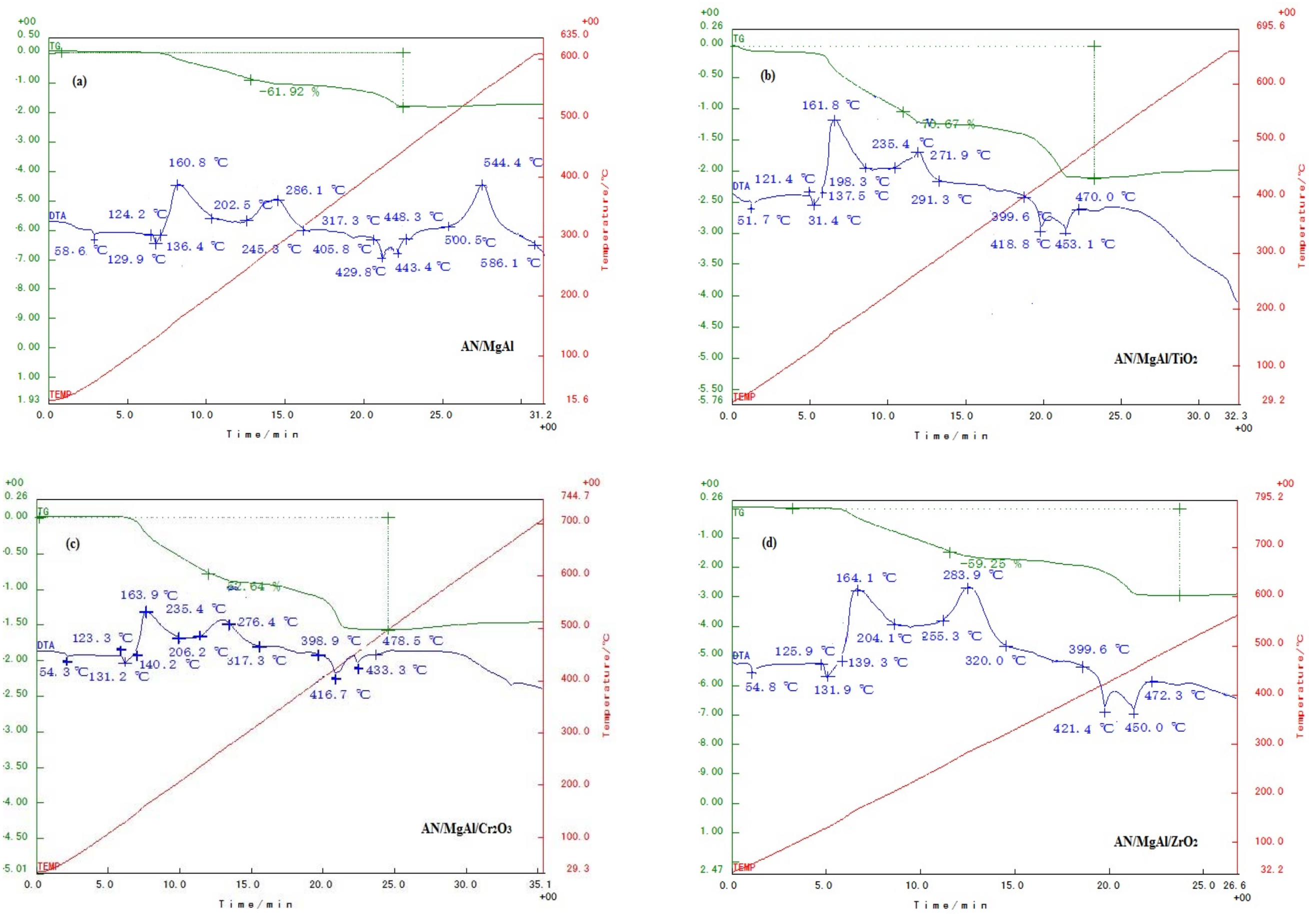


Fig. 5. The results of thermogravimetric and differential thermal analysis of the AN/MgAl and AN/MgAl/MeOx – based composite gas generators.

mass loss process took place in the temperature range from 245.3 °C to 405.8 °C. The previous loss of mass may be due to thermal decomposition of ammonium nitrate. And the second loss of weight may be caused by the release of oxygen in the reaction between ammonium nitrate and mechanical MgAl alloy. These two processes are exothermic. The two endothermic peaks on TDA line at the temperatures 429.8 °C and

443.4 °C correspond to the melting point of mechanical MgAl alloy and diffusion point of the gaseous phase to the liquid surface. Besides a little growth in TG line is observed with the appearance of the exothermic peak in the temperature range from 500.5 °C to 586.2 °C. Here, TG line stabilized the vapor phase reaction of NH_4NO_3 with liquid MgAl alloy complemented with metal oxide.

$$NH_4NO_3 + Mg \rightarrow MgO + N_2 + 2H_2O \quad (1)$$

$$3NH_4NO_3 + 2Al \rightarrow Al_2O_3 + 3N_2 + 6H_2O \quad (2)$$

As shown in Fig. 5b-d addition of transition metal oxides AN/MgAl-based composite gas generator does not change the general reaction to the temperature 443.8 °C. But, there was a change in peak shapes at the peak temperature and under thermal effects. In comparison with Fig. 4a in metal oxide gas generators, the temperature of the endothermic melting of MgAl alloy moves to lower values. We can assume that metal oxide shows improve burning characteristics in the gas-liquid condensed phase. Besides, a high reaction does not proceed with the diffusion of solid catalyst of transition metal oxides, MgAl alloy and NH_4NO_3 in the gaseous phase to the surface. Analyzing the obtained results, the gas generators are capable of the constant burning and have the ability to allocate large quantities of energy. So, combustion of gas generators is combustion of pure natural products. The use of metal alloys as a fuel compared to the use of pure metals allows the use of properties such as high energy, the lower ignition point, low density and low oxidation. Therefore, the use of mechanical MgAl alloy is considered to be promising as fuel in gas generators on the basis of ammonium nitrate. However, this study was conducted at low pressures and it is necessary to carry out researches at high pressure and study the different ratio of metal alloy with ammonium nitrate. Also investigated the use of various metal oxides as improve burning characteristics on gas generators.

4. Conclusions

The burning characteristics of the AN/MgAl – based gas generators were investigated based to improve burning characteristics of metal oxides. According to the obtained results, in the combustion chamber the lower flammability limit decreased from 2 MPa to1 MPa with addition of metal oxides to AN/MgAl – based gas generators. Also, the increase in the burning rate depending on pressure was shown. According to the results of the TG-DTA analysis, addition of mechanical MgAl alloy to ammonium nitrate lowered the temperature of thermodynamic aspects. But metal oxides did not significantly affect thermal decomposition of ammonium nitrate. However, the melting point of mechanical MgAl alloy was shown to decrease by 25 °C under to improve burning characteristics of metal oxides in the composition of AN/MgAl/MeOx system in the gas – liquid condensed phase.

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